Quantum and classical dynamics in complex one-dimensional antiferromagnets.

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Of great recent interest in condensed matter physics are phenomena of coexistence of quantum and classical properties in the same material. Such duality occurs in certain mixed-spin antiferromagnets composed of quantum spin chains interacting through "auxiliary" magnetic ions. In this category are linear-chain rare earth nickelates that exhibit a seemingly paradoxical co-existence of long-range magnetic order and one-dimensional gapped quantum spin excitations at low temperatures. In the present paper we propose a unified physical description of these compounds, supported by recent neutron diffraction and inelastic scattering studies. Our basic concept is the effective separation between low-frequency classical and high-frequency quantum spin correlations. This interpretation enables experimental measurements of some previously inaccessible fundamental properties of quantum spin chains, and predicts new exotic magnetic excitations and mechanisms of long-range ordering in complex quantum magnets.

CONTENTS

I. INTRODUCTION

For the past two decades one-dimensional (1D) quantum antiferromagnets (AFs) have been among the hot topics in condensed matter physics. Conventional three-dimensional magnetic materials exhibit long-range magnetic order (LRO) at low temperatures. In contrast, in one- and two-dimensional systems thermal fluctuations destroy LRO at any temperature $T > 0$. The 1D AF case is particularly interesting since even at $T = 0$ LRO is destroyed by *zero-point quantum fluctuations*. A breakthrough in understanding quantum AFs came with the theoretical work of Haldane (Haldane, 1983), who showed that spin correlations are radically different in integer- and half-integer 1D spin systems. Half-integer-spin chains are known to be in a *quantum critical state*, where spin correlations decay with distance according to a power law, and the excitation spectrum is gapless. In contrast, the ground state of integer spin chains is a "quantum spin liquid", with exponentially decaying correlations and an energy gap Δ in the magnetic excitation spectrum.

The wealth of experimental and theoretical results accumulated to date adds up to a fairly complete understanding of many of the properties of idealized 1D quantum AFs. One remaining problem of enormous interest is understanding *quasi*-1D systems: interacting quantum spin chains as part of *real* 3-dimensional crystal structures. Significant insight in this field has been obtained in studies of simple compounds with *directly coupled* quantum spin chains, such as $CsNiCl_3$ ($S = 1$) (see, for example, Buyers *et al.*,, 1986, Morra *et al.*, 1988, Enderle *et al.*, 1999, and references therein) and $KCuF_3$ ($S = 1/2$) (Tennant *et al.*, 1993, Tennant *et al.*, 1995, Lake *et al*, 2000, and references therein). Quasi-1D AFs are a particular case of the more general and very interesting phenomenon of the coexistence of quantum and classical spin fluctuations in the same material. In this context there have been a number of studies of classical systems with an added component of quantum mechanical interactions. An example of this is the realization of the so-called "Transverse Ising Model" recently found in certain holmium salts (Brooke *et al.*, 1999). In the present paper we discuss the opposite approach, starting from the quantum limit, and looking at quantum spin chains that interact with each other only indirectly, via an "auxiliary" network of essentially classical spins. The only known experimental realizations of this model are rare earth nickelates with the general formula R_2BaNiO_5, where R stands for a magnetic rare earth element. These "mixed" quantum-classical systems contain almost isotropic $S = 1$ AF Ni-chains weakly coupled

to magnetic rare earth ions. As discussed below, they exhibit an effective *separation* of classical and quantum spin dynamics in certain regimes. This provides a unique opportunity to investigate a number of interesting phenomena, including the effect of strong staggered magnetic fields on Haldane spin chains, and the peculiar interaction between collective quantum excitations in the chains and single-ion quantum transitions in the rare earth elements.

All R_2BaNiO_5 species are derivatives of Y_2BaNiO_5, an isomorphous compound in which the magnetic R^{3+} are replaced by non-magnetic Y^{3+} ions. Y_2BaNiO_5 is a textbook example of a Haldane-gap AF (Darriet and Regnault, 1993, Yokoo *et al.,* 1995, Xu *et al.*, 1996). Its magnetic properties are due to chains of $S = 1$ Ni^{2+} ions that run along the a axis of the orthorhombic crystal structure (Fig. 1). The Ni-spins are coupled by rather strong ($J \sim 300$ K) AF nearest-neighbor intra-chain interactions. Inter-chain coupling is negligible. Down to very low temperatures this system shows no sign of magnetic ordering and has a thermally-activated magnetic susceptibility: a signature of a spin-singlet ground state and energy gap. Haldane-gap excitations in Y_2BaNiO_5 ($\Delta \approx 10$ meV) have been observed and extensively studied in inelastic neutron scattering experiments (Darriet and Regnault, 1993, Yokoo *et al.*, 1995, Xu *et al.*, 1996). In R_2BaNiO_5 compounds, the place of Y^{3+} is taken by magnetic rare earth ions ranging from Pr^{3+} to Tm^{3+} (GarciaGarcía-Matres *et al.*, 1993, GarciaGarcía-Matres *et al.*, 1995). Even though direct interactions between the R^{3+} moments are negligible, their coupling to the Ni-chains in all cases leads to long-range Néel ordering with transition temperatures T_N ranging from 24 K (Pr_2BaNiO_5) to 65 K (Dy_2BaNiO_5). A static ordered moment appears on both the Ni- and R-sites simultaneously upon cooling through T_N (Figs. 2a,b). At a first glance it may seem that the magnetic LRO is a clear sign of a destruction of the singlet ground state in individual Haldane chains. Indeed, for quite some time R_2BaNiO_5 compounds were considered to be classical magnets.

A breakthrough came when, in inelastic neutron scattering experiments on Pr_2BaNiO_5 (Zheludev *et al.*, 1996-1), Nd_2BaNiO_5(Zheludev *et al.*, 1996-2, Yokoo *et al.*, 1998, Raymond *et al.*, 1999, Zheludev *et al.*, 2000), and $(Nd_xY_{1-x})_2BaNiO_5$ (Yokoo *et al.*, 1997, Yokoo *et al.* , 1998), it was discovered that 1D Ni-chain gap excitations, strikingly similar to the Haldane modes in Y_2BaNiO_5, not only exist above the ordering temperature, but also *persist in the magnetically ordered state*. One could argue that these excitations in R_2BaNiO_5 are classical spin waves, that acquire a gap due to magnetic anisotropy effects, and that the similarity with Y_2BaNiO_5 is coincidental. This interpretation, however, is clearly inappropriate. The gap energy (around 10 meV) is too large to be attributed to anisotropy on the Ni-sites (less than 1 meV, according to Xu *al.*, 1996). At the same time, it can not be attributed to anisotropy involving the rare earth ions either, being independent of the type of rare earth involved, and even remaining unchanged in the entire concentration range of mixed Y-R compounds. Another hint is that classical spin waves represent the spontaneous breaking of $O(3)$ symmetry in the magnetically ordered state, and are thus a doublet of excitations. In contrast, gap excitations in R_2BaNiO_5 were shown to be a *triplet* (Raymond *et al.*, 1999), just like Haldane excitations from an $O(3)$-invariant spin liquid state. One is forced to conclude that the 1D quantum spin correlations in the Haldane chains somehow survive the onset of LRO in R_2BaNiO_5 compounds.

II. TWO FREQUENCY REGIMES

The key to resolving this apparent contradiction lies in the very short time scale associated with dynamic spin correlations in individual chains, which in turn is a *direct consequence of the energy gap*. At frequencies substantially smaller than the threshold frequency $\Delta/\hbar$, the dynamic susceptibility $\chi(\omega)$ of an isolated $S = 1$ AF chain is purely real and is *only weakly frequency-dependent*. In R_2BaNiO_5 the magnitude $J_\perp$ of Ni-R exchange interactions is typical of a rare-earth-transition-metal superexchange bond, and is of the order of 1 meV. This energy and corresponding frequency scales are much smaller than the Haldane gap Δ (10 meV). At such frequencies the local magnetization of the Ni-chains is able to instantaneously follow any fluctuations of neighboring rare earth spins. The effect of the quantum spin chains is then reduced to simply providing an effective pathway for R-R interactions of the magnitude $J_{\rm eff} \sim J_\perp^2 \chi(0)$. We end up with a 3D network of essentially classical rare earth moments that, not surprisingly, orders magnetically at low temperatures. The low-frequency dynamics are then also that of a classical magnet. Conversely, the same arguments suggest that the dynamic spin correlations in the high-frequency range are those of a 1-D $S = 1$ quantum AF in a *static* effective field generated by the rare earth subsystem. In other words, in the limit of weak coupling, for two-component systems that include *gapped* quantum spin chains there is a *separation* between static and low-frequency properties (LRO and classical spin waves) and the high-frequency behavior (quantum 1-D excitations). In the next two sections, we shall review the experimental and theoretical results that support this physical picture.

III. STATIC PROPERTIES

A quantitative model that embodies the hand-waving arguments given above is the chain-mean-field (chain-MF) theory (Scalapino *et al*, 1975, Affleck, 1989). The

standard MF approach for conventional magnets starts out with the bare (non-interacting) susceptibilities for all individual magnetic ions involved. In the chain-MF theory, one respects the fact that Ni-R coupling is substantially smaller than the strong interactions within the Ni-chains themselves. The chains are thus treated as single entities. For the R_2BaNiO_5 series the low-temperature magnetic structure is antiferromagnetic, so the relevant properties of the spin chains are their *staggered magnetization function* $M_\pi(H_\pi)$ and *staggered susceptibility* $\chi_\pi = dM_\pi/dH_\pi$. As for the rare earth ions, for those with a Kramers doublet ground state configuration (Nd^{3+} and Er^{3+} among them) it is appropriate to take the standard approach and write the non-interacting response in the form of a Brillouin function for an isolated ion. The coupling between these two systems is then treated at the MF level and all static properties, including long-range ordering and the T-dependence of the magnetic order parameters are derived from a set of self-consistent equations. The problem is that to solve these equations one has to know $M_\pi(H_\pi)$ for an isolated chain. Prior to the experimental studies of R_2BaNiO_5 this function was not known analytically or even numerically. Simple powder-diffraction measurements on $(Nd_xY_{1-x})_2BaNiO_5$, Er_2BaNiO_5 and Ho_2BaNiO_5 however, allowed the direct *measurement* of this fundamental property (Zheludev *et al.*, 1998, Yokoo *et al.*, 1998). The magnetic ordering temperatures (around 50 K) are considerably smaller than the Haldane gap energy ($\Delta/k_B \sim 120$ K) and, in this regime, $M_\pi(H_\pi)$ is almost T-independent (Kim, 1998). The effective staggered field acting on the spin chains in R_2BaNiO_5 is proportional to the rare earth magnetic order parameter. From these two facts it follows that $M_\pi(H_\pi)$ may be obtained simply by plotting the measured ordered moment on the Ni-sites vs. that on the R-sites, as shown in Fig. 2(c). The scaling of the abscissa depends on the MF coupling constant and obviously on the type of rare earth ions involved. Universal scaling in proper magnetic field units is achieved by substituting the measured $M_\pi(H_\pi)$ curve into the self-consistent MF equations. The MF coupling constants are then refined by fitting the measured temperature dependencies of sublattice magnetizations (Zheludev *et al.*, 1998, Yokoo *et al.*, 1998). Excellent fits can be obtained in this manner (Fig. 2a,b-solid lines). The resulting field scale is shown in the top axis of Fig. 2(c). The data collapse for systems with substantially different ordering temperatures and saturation moments is quite impressive, validating this approach. Note that staggered fields of up to 40 T are being produced in these experiments!

It is important to realize that the mechanism of magnetic ordering in R_2BaNiO_5 materials is quite different from that in systems with directly coupled chains, such as $CsNiCl_3$. In the latter, LRO results from a complete softening of the Haldane excitations at the 3-D AF zone-center as $T \to T_N$. This can happen only if inter-chain interactions exceed some critical value (Affleck, 1989). In the case of rare earth nickelates, magnetic ordering is expected to occur for any *arbitrarily small* Ni-R coupling, as a result of the $1/T$-divergence in the bare susceptibility of individual rare earth moments.

IV. SPIN DYNAMICS

While long-range magnetic ordering is beautifully described by the chain-MF model, the spin *dynamics* in our two-component magnets can be analyzed with the chain-Random-Phase-Approximation (RPA). At this level, just as was the case for the static properties, the low-energy magnetic excitations in R_2BaNiO_5 can be described in terms of effectively coupled classical rare earth spins. Such classical spin waves were recently studied in Nd_2BaNiO_5 (Yokoo *et al.*, 1998). The Nd-case is particularly simple because of a strong easy-axis magnetic anisotropy associated with Nd^{3+}. The spin waves are then dispersionless and resemble excitations in a conventional Ising magnet. The excitation energy is given by $2M_{Nd}H_\pi$, where M_{Nd} is the saturation moment of Nd^{3+} and H_π is the effective mean field produced by the Ni-sublattice. In fact, the measured temperature dependence of this energy can be accurately reproduced, without using any adjustable parameters, by utilizing $H_\pi(T)$ determined from the MF-analysis of the sublattice magnetization data.

We now turn to the Ni-chain gap excitations. In the paramagnetic phase (above T_N) there is no ordered moment on the rare earth sublattice, so the 1-D gap modes are exactly as those in uncoupled chains. Thus in Nd_2BaNiO_5 and Pr_2BaNiO_5, for $T > T_N$, the 1-D gap excitations are experimentally identical to those found in Y_2BaNiO_5 (Zheludev *et al.*, 1996-1), Nd_2BaNiO_5(Zheludev *et al.*, 1996-2, Yokoo *et al.*, 1998, Raymond *et al.*, 1999). In particular, the T-dependence of the gap energy is very similar (Fig. 3a). Within the framework of our MF-RPA model, in the magnetically *ordered* state, the gap excitations are those of isolated quantum spin chains immersed in a *static* staggered exchange field. Experimentally, Haldane-gap modes are indeed observed for $T < T_N$ in all R_2BaNiO_5 materials studied to date. The striking experimental result is that the gap energy *increases* with decreasing T below T_N, roughly linearly with $T - T_N$ (Fig. 3a). Moreover, the behavior appears to be independent of the type of rare earth involved or the actual Néel temperature. The best way to demonstrate this universality is to eliminate the rare-earth related energy scale completely by plotting the increase of the Haldane gap, normalized to that in Y_2BaNiO_5, as a function of the induced static staggered moment on the Ni-sites (Fig. 3b). When plotted

in this way, the data obtained for different R_2BaNiO_5 systems collapses onto a single curve. This is a clear indication that an increase of the Haldane gap energy in the presence of a staggered field is an *intrinsic* property of the quantum spin chains.

Despite the simplicity of its origin, the effective separation of classical and quantum dynamics in these materials has a powerful consequence: it turns the R_2BaNiO_5 compounds into unique model systems for fundamental studies of quantum spin chains in strong staggered fields, *i.e.*, fields modulated on the *atomic* length scale. The direct measurements of the staggered magnetization curve $M_\pi(H_\pi)$ and the staggered field dependence of the Haldane gap $\Delta(H_\pi)$ described above, stimulated intensive theoretical studies. A particularly successful approach (Maslov and Zheludev, 1998, Yokoo *et al.*, 1998) is the $O(3)$-symmetric (1+1)-dimensional field theory (the so-called ϕ^4 model), first used by Affleck to describe Haldane spin chains (Affleck, 1989). In this theory it becomes apparent that the non-linearity of $M_\pi(H_\pi)$ and the monotonous behavior of $\Delta(M_\pi)$ are both a manifestation of *repulsion* between single-particle gap excitations. Coefficients characterizing this repulsion in a fully renormalized Hamiltonian can be estimated numerically and used to obtain power series expansions for $M_\pi(H_\pi)$ and $\Delta(M_\pi)$ (Maslov and Zheludev, 1998). These predictions are in excellent agreement with all of the R_2BaNiO_5 data and are shown as solid lines in Figs. 3b and Fig. 2c. The results obtained in neutron scattering experiments on R_2BaNiO_5 have also been confirmed by numerical simulations (Lou *et al.*, 1999, dashed line in Fig. 3b). Very recently, the increase of the gap energy in a two-component magnet was rigorously proven for the Valency Bond Solid model (Bose and Chattopadhyay, 1999), known to possess many similarities with 1-D $S = 1$ Heisenberg model relevant to the present case.

V. THE ROLE OF SINGLE-ION EXCITATIONS

Above we restricted ourselves to considering Haldane chains coupled to essentially classical spins. Qualitatively new phenomena occur when the "auxiliary" magnetic ions themselves have non-trivial intrinsic dynamics. Due to strong spin-orbit interactions and low site-symmetry, this in fact is always the case for R^{3+} in R_2BaNiO_5. Not just the ground state multiplets of the rare earth ions, but also their higher-energy excited crystal-field (CF) configurations will couple to the integer-spin chains.

One can identify two distinct regimes. The first regime is realized when the R-centered single-ion CF transitions occur close to, or above, the Haldane gap. CF excitations in this case have little influence on the low-frequency and static properties of the system, for which the classical model remains adequate. At high frequencies though, the proximity of single-ion modes on the R sites to 1-D excitations in the chains gives rise to peculiar mixed Ni-R modes. Such behavior was recently observed in inelastic neutron scattering experiments on Nd_2BaNiO_5(Zheludev *et al.*, 2000). Figures 4(a,b) show representative constant-Q scans collected at the 1-D AF zone-center for different momentum transfers perpendicular to the chain axis, at $T = 55$ K (*above* $T_N = 48$ K). Three separate peaks, namely the Haldane-gap excitation at 11 meV energy transfer and two CF excitations, a weaker mode at 18 meV and a strong one at 24 meV, can be clearly identified. A striking feature is the strong variation of the intensity of these modes as a function of transverse momentum transfer, as shown in (Fig. 4c). This intensity modulation results from an *interference* between Ni- and Nd- spin fluctuations. The gap excitations retain their predominantly 1-D character, and can be described as Haldane modes propagating on the Ni-chains, "dressed" by correlated CF fluctuations on the Nd-sites. This behavior is well accounted for by a slightly more sophisticated version of the chain-RPA model, that includes higher-energy levels of the rare earth ions. It is interesting that at the 1-D AF zone-center the interference effect barely influences the energies of the modes coupled. Only a very weak transverse dispersion is seen in the Haldane branch (Fig. 4d), and none can be detected in the CF excitations within experimental energy resolution. Our interpretation of the T-dependence of the gap in terms of Haldane chains in a static staggered field is thus justified. It is also clear that the Ni-Nd interactions in Nd_2BaNiO_5 are too weak to drive the Haldane gap to zero energy at any wave vector, which would be required to produce a $CsNiCl_3$-like transition to a Néel-like state (Buyers *et al.*, 1986, Morra *et al.*, 1988, Affleck, 1989).

Away from the 1-D AF zone-center the mixing between Haldane excitations and local modes can be quite dramatic in the isolated regions of reciprocal space where their non-interacting energies coincide. Here both intensities and energies of coupled modes are severely affected. In fact, the distinction between single-ion and chain modes is totally blurred: the former continuously "morph" into the latter, and vise versa. This "anticrossing" phenomenon in Nd_2BaNiO_5 is illustrated in Fig. 5, that shows a mesh composed of data points collected in constant-E scans along the chain axis.

Quite different is the second regime, where the energies of certain excited CF states are considerably smaller than the Haldane gap. As in the case of classical "auxhiliary" spins, rare earth- and Ni-chain- spin dynamics are effectively separated. At low frequencies the Haldane chains simply provide effective pathways for direct R-R interactions. This effective coupling becomes particularly important for non-Kramers rare earth ions that in the structure of R_2BaNiO_5 have a non-magnetic ground state. A good example is Pr_2BaNiO_5, where Pr^{3+} has a spin-singlet ground state, but in addition has a CF-

excited state at only 4 meV energy. Above $T = 100$ K the corresponding CF mode is dispersionless. With decreasing temperature it acquires a substantial dispersion and intensity variation both along, and perpendicular to, the chain axis (Zheludev *et al.*, 1996-1). With two Pr sites per every Ni-site, The CF mode becomes split into an optic and accoustic branches. As shown in Fig. 6, the accoustic branch develops a sharp dip at the 3D AF zone-center, where its energy reaches zero at $T_N = 24$ K. A magnetic *soft-mode* transition occurs at this point, and is driven by Ni-chain-mediated Pr-Pr interactions. This soft-mode behavior is very similar to that in metallic Pr (for a good review of the subject, see book by Jensen and Mackintosh). Below T_N Pr_2BaNiO_5 is ordered in a structure similar to that of Nd_2BaNiO_5. In an unusual twist, magnetic LRO in this case is a result of interaction between two *non-magnetic* systems: singlet-ground-state rare-earth ions and quantum-disordered spin chains.

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In summary, complex systems composed of gapped quantum spin chains interacting through isolated free spins demonstrate a rich spectrum of magnetic properties with both classical and quantum-mechanical features. R_2BaNiO_5 rare earth nickelates are unique model compounds in which these phenomena can be studied experimentally.

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FIG. 1. A schematic view of the R_2BaNiO_5 crystal structure. $S = 1$ Haldane spin chains are formed by Ni^{2+} ions at the centers of NiO_6 octahedra , arranged along the a crystallographic axis. The rare earth ions provide magnetic links between chains.

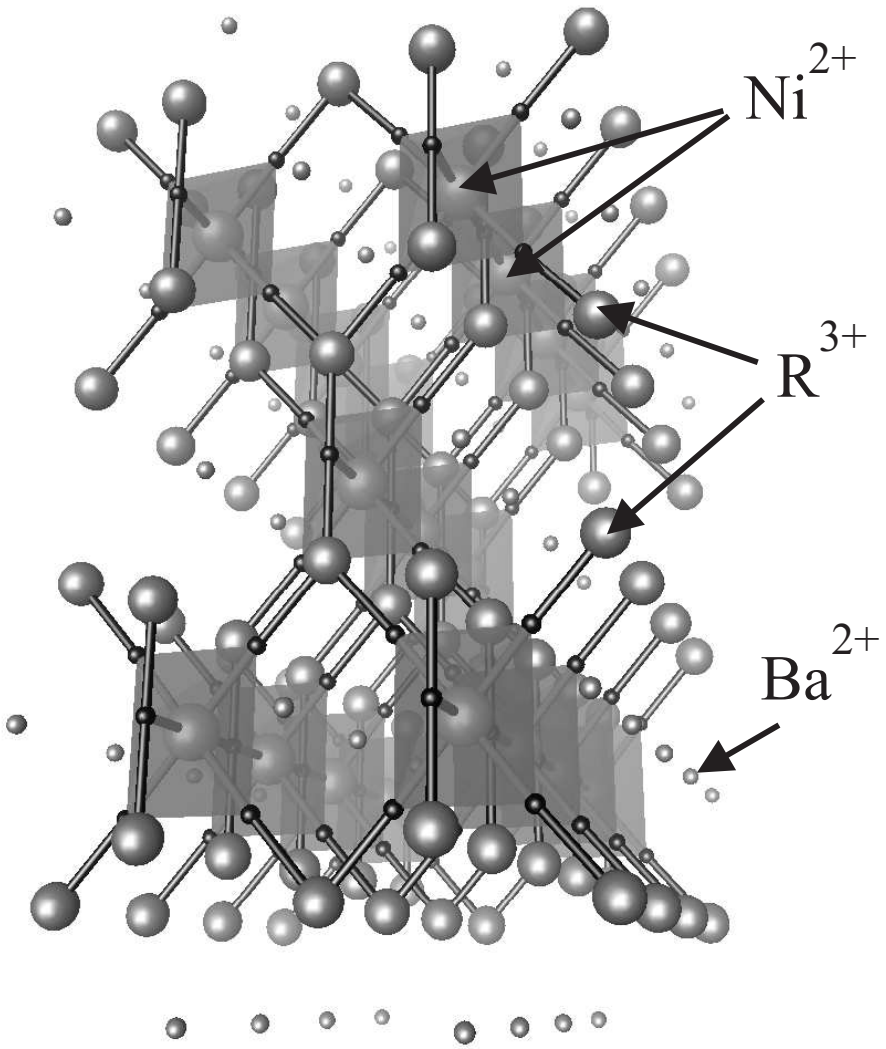
FIG. 2. (a) Temperature dependence of the Ni- and R-sublattice magnetizations measured in Nd_2BaNiO_5 (symbols) (Yokoo *et al.*, 1998) using neutron powder diffraction. The solid lines are chain-MF theoretical fits. (b) Same for Er_2BaNiO_5(Alonso *et al.*, 1990). (c) Measured induced staggered moment on the Ni-sites (symbols) plotted against the magnetic order parameter of the R-sublattice in several R_2BaNiO_5 compounds (bottom axes). The collapsed data sets can be interpreted as a measurement of the universal staggered magnetization curve for Haldane spin chains (top axis). The solid line is a theoretical prediction based on the ϕ^4 model.

FIG. 3. (a) Measured temperature dependence of the Haldane gap energy for a series of Y-substituted Nd_2BaNiO_5 compounds (symbols), as measured by inelastic neutron scattering. Above the corresponding temperatures of magnetic ordering all systems show the same behavior as the intrinsically disordered Y_2BaNiO_5 (Yokoo *et al.*, 995) . In the ordered phase the gap increases in all cases linearly with T. Lines are guides for the eye. (b) Data collapse obtained by plotting the measured difference between the gap in $(Nd_xY_{1-x})_2BaNiO_5$ and that in Y_2BaNiO_5 as a function of the induced staggered moment on the Ni-chains. The solid line is a parameter-free prediction of the ϕ^4 model, and the dashed line represents recent numerical results (Lou *et al.*, 1999) for Haldane chains in a staggered field.

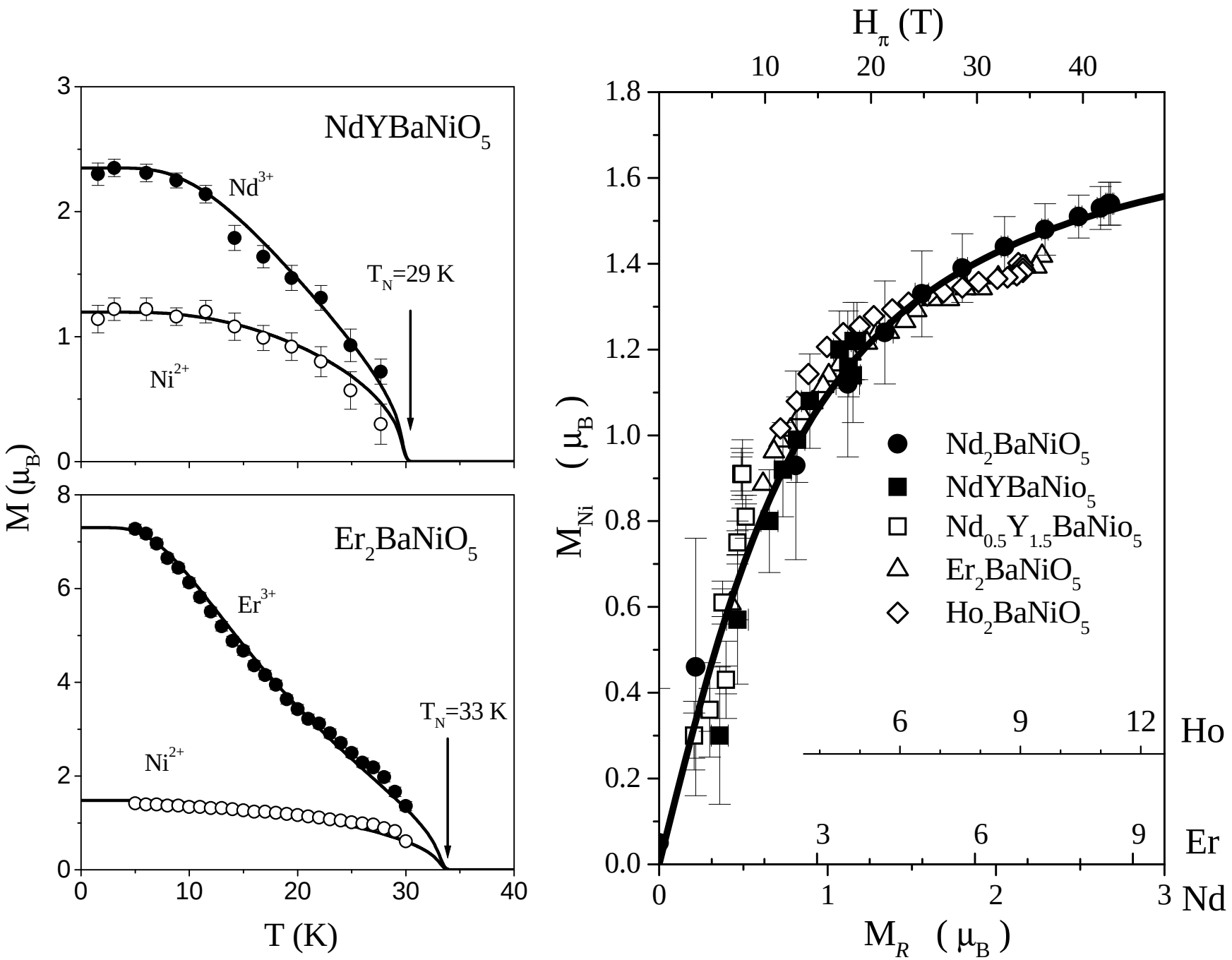
FIG. 4. Mixing between Haldane-gap and crystal field excitations in Nd_2BaNiO_5, observed in single-crystal inelastic neutron scattering experiments. (a,b) Typical constant-Q scans measured at the 1D AF zone-center for two different momentum transfers perpendicular the chain-axis (symbols). The data were taken on the HB-1 3-axis spectrometer at Oak Ridge National Laboratory, using $E_f = 14.7$ meV fixed-final energy neutrons, pyrolitic graphite (PG) monochromator and analyzer, $60' - 80' - 80' - 240'$ collimations and a PG filter after the sample. The solid black line is a fit to a semi-empirical model cross section taking into account resolution effects. The red, green and blue peaks represent the Haldane and two CF excitations, respectively. This analysis reveals a substantial intensity modulation in all modes (c) and a weak dispersion in the Haldane mode (d).

FIG. 5. Grayscale/contour plot of inelastic neutron scattering intensity measured in Nd_2BaNiO_5 at the 1D AF zone-center shows the highly dispersive Haldane-gap mode (dashed parabola) and the flat 24 meV crystal field mode (dashed horizontal line). The data were taken using the same setup as for Fig. 4. The symbols and attached bars show the positions and widths of peaks seen in constant-E (circles) and constant-Q (squares) scans, respectively. A mixing between the two modes leads to an anticrossing effect (solid lines).

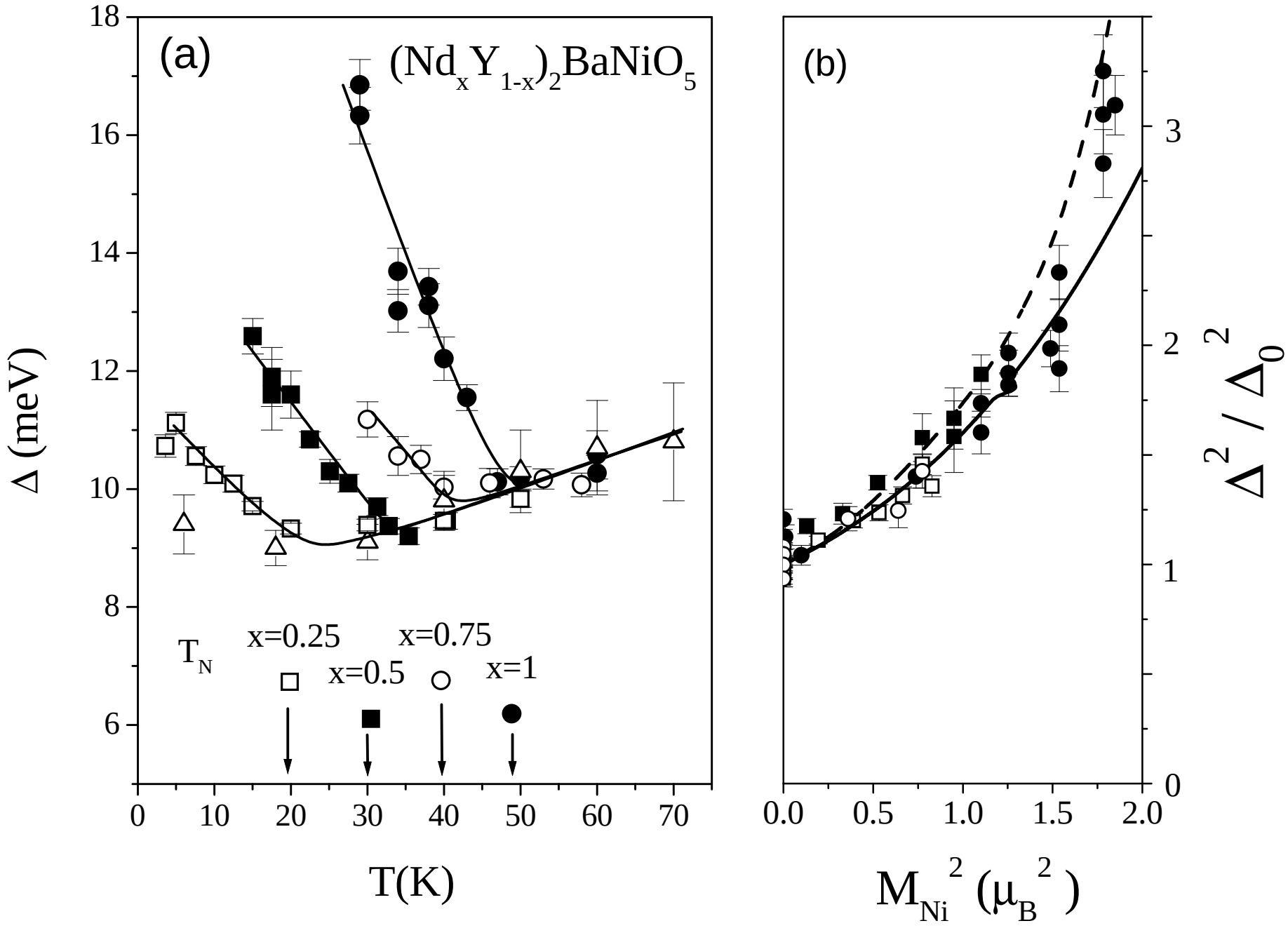
FIG. 6. Temperature dependence of the 3D AF zone-center excitation energies measured in Pr_2BaNiO_5. A repulsion between single-ion Pr CF excitations (circles) and Haldane-gap modes in the Ni-chains (triangles) leads to a soft-mode transition to a magnetically ordered state below $T_N = 24$ K, despite the singlet (non-magnetic) nature of the Pr^{3+} ions. The Haldane-gap modes behave very similarly to those in Nd_2BaNiO_5.



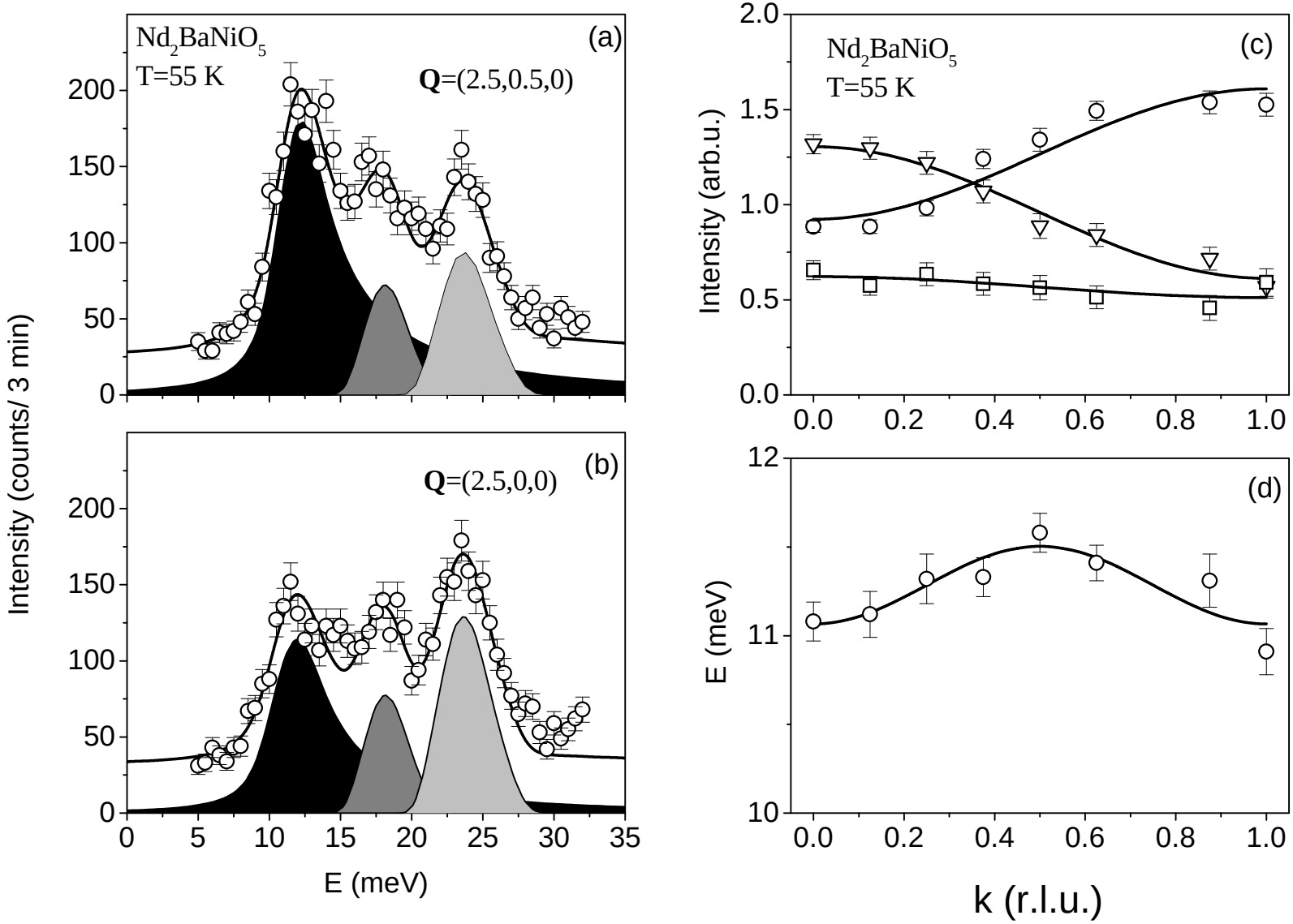
Zheludev et al. Fig. 1



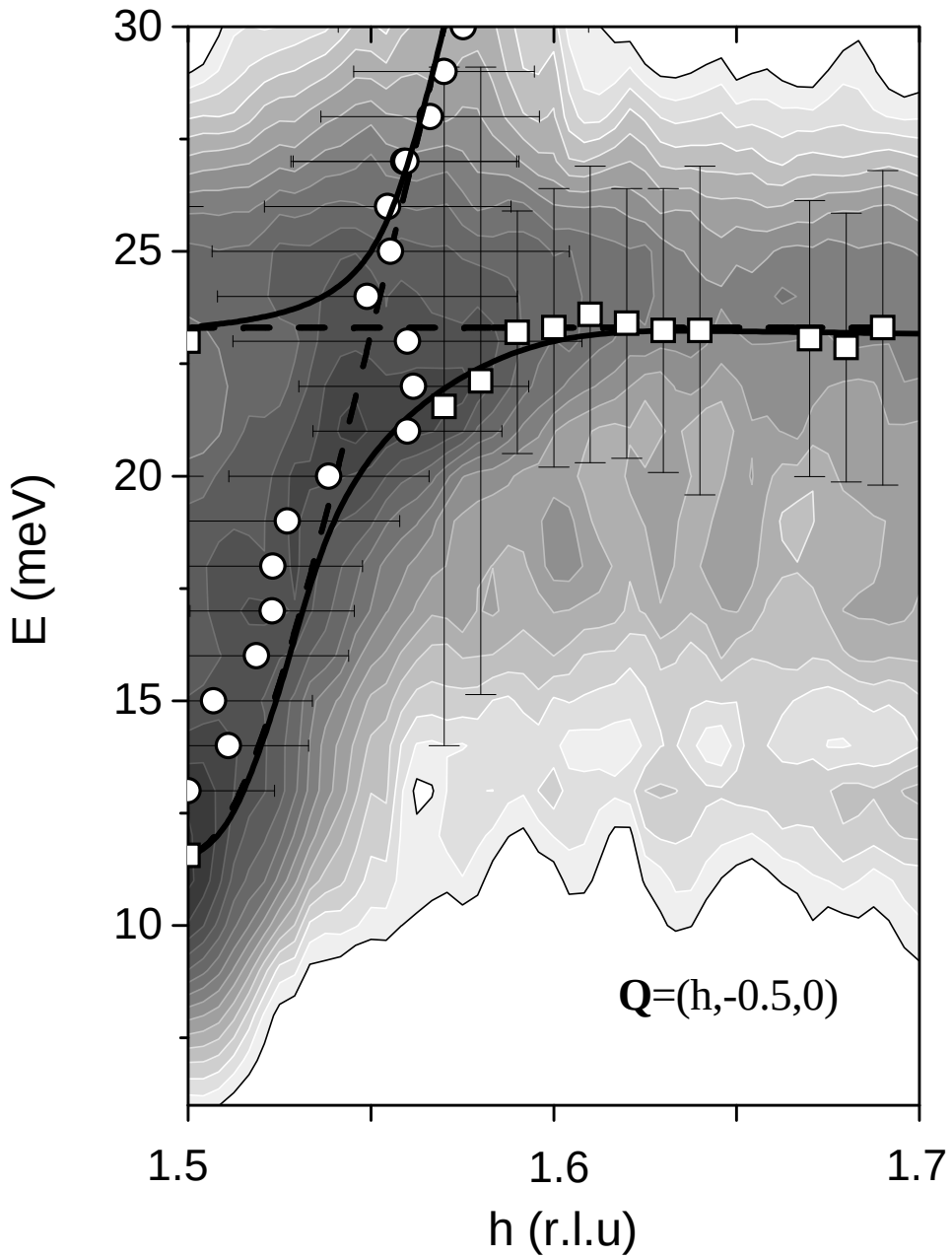
Zheludev et al. Fig. 2



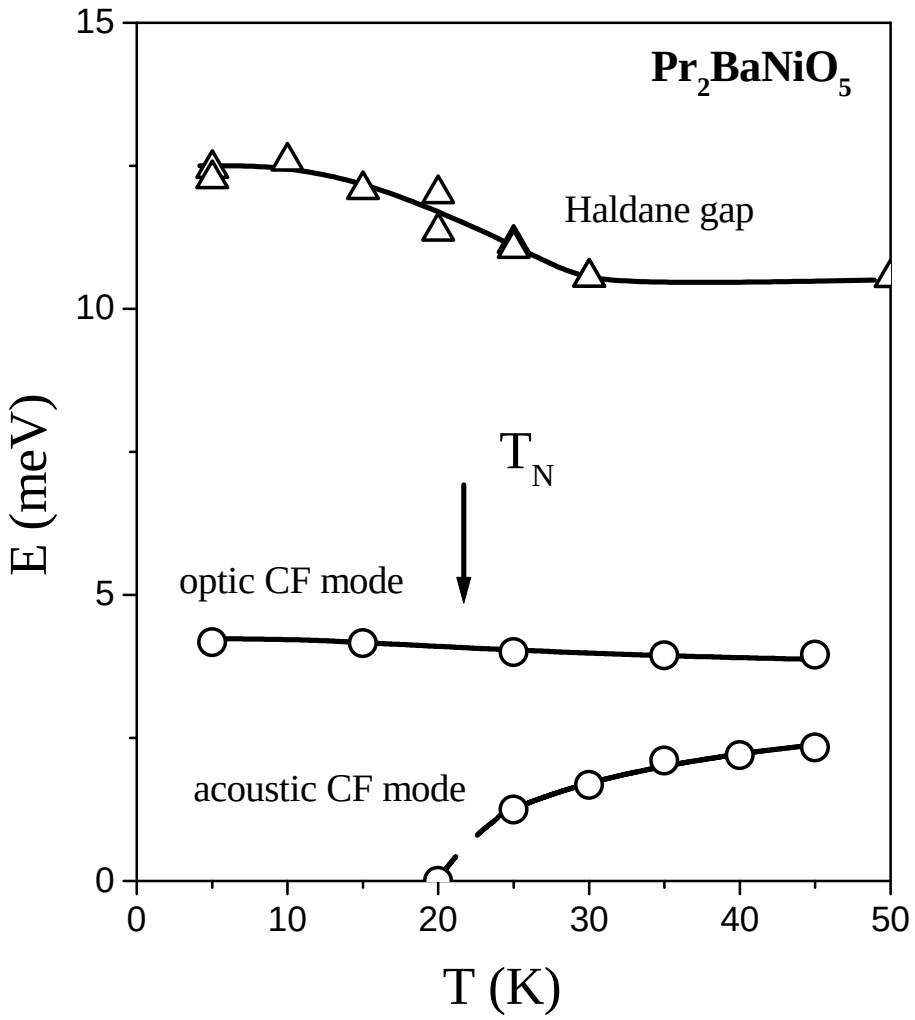
Zheludev et al. Fig. 3



Zheludev et al. Fig. 4



Zheludev et al. Fig. 5



Zheludev et al. Fig. 6